Spectral Signature of the Kondo Lattice in Angle-Resolved Photoemission of URu_2Si_2

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INTRODUCTION

Heavy fermion compounds are characterized by a large linear specific heat coefficient (γ) which signals large ('heavy') effective masses of electrons forming the Fermi surface [1]. Resonant photoemission spectroscopy of polycrystalline samples has contributed to the understanding of rare earth and actinide heavy fermions by identification of the *f*-electron weight contribution to the states at E_F. For many rare earth systems this *f*-weight can be analyzed and related to γ within a single impurity Kondo picture [2, 3]. Nonetheless from general theoretical ideas [4] and early experimental de Haas-van Alphen studies [5] it has been appreciated for very many years that this *f*-weight should be part of the dispersions which define the Fermi surface.

The focus of this work is the detailed ***k***-dependent *f*-electronic structure in the heavy fermion compound URu_2Si_2, which has a moderately large linear specific heat coefficient of γ~180 mJ/mol-K^2 [6] extrapolated from the paramagnetic phase (T>17.5 K). By combining angle-resolved photoemission (ARPES) and resonant photoemission on single crystal surfaces, the *f* and *d* contribution to the dispersing bands can be distinguished and compared to theoretical models of heavy-mass formation at the Fermi surface. Initial ARPES experiments on URu_2Si_2 zone [7, 8] have (a) determined the crystal inner potential (V_0~12 eV), (b) mapped the basic *d*-band structure along high symmetry directions and (c) established the existence of hole pockets at the Γ, Z and X-points of the Brillouin. Good agreement between the experimental *d*-band structure and theoretical band calculations [9, 10] is observed. However, near-E_F, the disagreement is severe due to correlation effects and *f-d* hybridization. This abstract focuses on these effects at the X-point in URu_2Si_2.

EXPERIMENT

URu_2Si_2 has the $ThCr_2Si_2$ crystal structure with a body-centered tetragonal Brillouin zone. Single crystal samples were cleaved in ultra-high vacuum (1.2×10^{-10} torr) at room temperature exposing the [001] surface and then cooled to ~150 K for ARPES measurements at ALS Beamline 7.0.1.2. The Fermi-edge intensity mapping technique using automated angular motions was employed to gain an overview of the sample orientation and Fermi surface structure. Photon energies above and below the U 5*d*→5*f* absorption thresholds (98 and 108 eV) were used to compare spectra dominated by *d*-band spectral weight to spectra with U 5*f* weight resonantly enhanced. A total instrumental resolution of ≈80 meV and full angular acceptance of ≈1.4° was employed.

RESULTS

Figure 1 shows the detailed electronic structure near the X-point in URu_2Si_2 below and above the the U 5*f* resonances including Femi-edge intensity maps (a,b,e) and dispersing valence band spectra (c,d). Below resonance at 85 eV where *d*-character is expected to dominate, a closed Fermi-surface topology is observed in the angular map in Fig. 1(a). Above resonance at 112 eV

where U 5*f* weight is enhanced, the interior of the hole-surface is observed to fill-in with intensity in Fig. 1(b). Spectra taken along the diagonal line shown in Fig. 1(a) indicate a clear hole-like dispersion present at both photon energies [Fig. 1(c,d)] creating this piece of Fermi surface. Consistent with the E_F-intensity maps, above resonance a sharp E_F peak which we attribute to U 5*f* weight is enhanced in the interior of the hole-pocket.

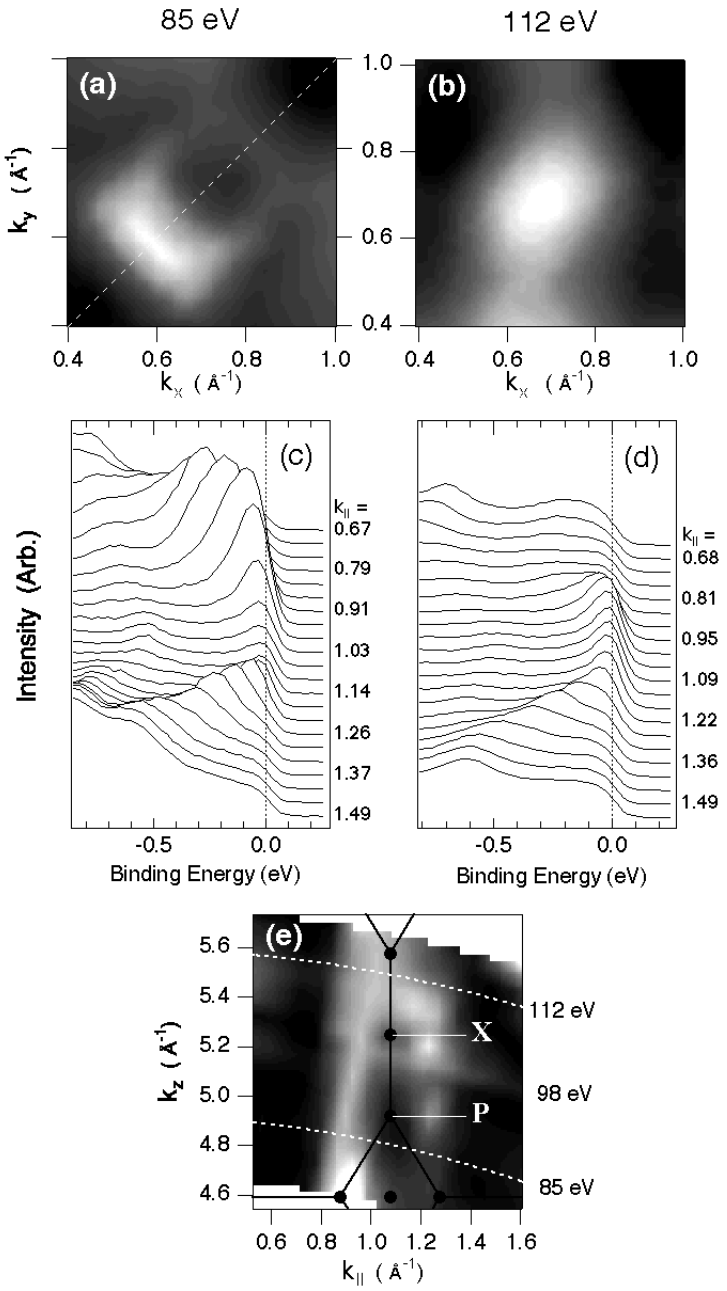

Figure 1. (a,b) Zoomed k_X-k_Y E_F-intensity maps of the X-point at 85 and 112 eV. (c,d) Spectra taken along the diagonal trajectory showing hole-like dispersions. (e) A k_X-k_Z E_F-intensity map, normalized to the maximum intensity for each photon energy, showing the FS topology along the P-X-P line and the filling in of the hole-pocket with *f*-weight above resonance. Dashed arcs indicate the trajectories of the on- and off-resonance spectra.

To address whether this change from below to above resonance is due to different k_Z locations within the 3-dimensional Brillouin zone and not merely due to varying *d* and *f* cross sections, the k_Z-dependence of the X-point was explored by an E_F-intensity map acquired by varying the photon energy and one angle of the sample. Fig. 1(e) was shows this k_X-k_Z map corresponding to a vertical slice in k-space with the Brillouin zone boundaries overplotted. This figure illustrates that the k-parallel value termed the 'X'-point so far is actually a vertical -P-X-P- line and that the maps and spectra acquired at 85 and 112 eV are observed to cut close to the P-points, i.e. similar locations of the Brillouin zone. The k_X-k_Z image, which has been normalized by an angle-integrated intensity profile in order to remove the strong overall f-resonance and highlight relative intensities, shows that this hole-surface extends along the P-X-P line. The relative intensity in the interior of this extended hole-surface is thus observed in Fig. 1(e) to increase at 98 eV and above 108 eV, confirming this effect to be due to the U 5*f* cross section enhancement at the U 5*d*→5*f* edges.

ANALYSIS

A hole-surface extended along the P-X-P line is very suggestive of a two-dimensional surface state with no dispersion along k_Z. In addition, no such hole-surface is predicted in theoretical band calculations. However, the persistence of this hole surface with surface contamination is equally robust with other bulk features. While theoretical LDA band calculations do not predict a hole-surface at the X-point, they do predict minimal dispersion along the P-X-P k_Z-line which repeats twice as fast as Γ–Z–Γ along the Brillouin zone centers. This symmetry property along P-X-P coupled with experimental k_Z-broadening orginating from the finite inelastice mean free path of photoelectrons can easily give the appearance of a two-dimensional state.

Regardless of the origin of the X-point Fermi-surface, the clear correlation of *f*-weight at E_F to the underlying *d*-band structure and Fermi surface, i.e. confinement to the center of a hole pocket, is observed for the first time in this data set on URu_2Si_2. This key effect, which goes beyond predictions of LDA band calculations, can be rationalized in the framework of Anderson lattice treatments [11-13]. In these many-body treatments, the bare *f*-level binding energy gets renormalized to just above E_F with renormalized hybridization to *d*-states in the vicinity of a *d*-band E_F crossing. The problem thus gets theoretically mapped onto a one-electron two-band model where hybridization between a flat *f*-band and a dispersing *d*-band opens up a gap at the renormalized *f*-energy and pushes *f*-weight down towards E_F on the unoccupied side of the *d*-band crossing (corresponding to the interior of a hole pocket). Hence the observation of enhanced *f*-weight at the interior of a hole-pocket is a generic signature of the many body Kondo lattice.

In addition the *d-f* character varies continuously along the two branches of the hybridized dispersions and the Fermi velocity of the original *d*-band gets reduced indicating the formation of a heavier mass FS with *f*-character. ARPES experiments at lower temperature and higher energy and angular resolutions are currently being performed in order to quantify the intrinsic spectral lineshapes at this heavy mass crossing arising from *f-d* mixing.

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